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A STUDY OF DEVELOPMENT DEFECTS AND TRACK STRUCTURES IN NUCLEAR EMULSIONS USING AMIDOL DEVELOPERS

O. N. KAUL

SAHA INSTITUTE OF NUCLEAR PHYSICS, CALCUTTA.

(*Received, December 30, 1960*)

ABSTRACT. Development defects in nuclear emulsions, and also the dependence of track and grain structures on the various development parameters have been investigated.

Modified formulae have been suggested for the removal of microscopic fog, coloration defects, and also for the background eradication.

The procedures of investigation followed during the course of this work, differ from the ones adopted by others like Fatzer, Yagoda, Barschall, and Liebermann; and the results, although identical in most of the cases, differ slightly in certain respects as reported.

Some work has been reported on the elimination of development defects by various workers, e.g., Yagoda, (1948), Liebermann and Barschall (1943) and others. The present aim of the author has been to make a comprehensive study of all development defects introduced by Amidol developers to compare the methods suggested by various workers for their elimination and also to suggest modified formulae suitable for work in this laboratory.

Besides this, a critical study of the track and grain structures pertaining to their dependence on various development parameters (temperature, time, and sodium sulphite concentration) has been made by methods somewhat different (from those adopted by Fatzer (1959).

Ilford C_2 nuclear emulsions of 100, 200 and 400 micron thickness were used and the problem studied under the following heads :

(1) Elimination of the development defects introduced by Amidol developers.

(2) Dependence of track and grain structures on the hot stage temperature,, hot stage time and sodium sulphite concentration.

(3) Removal of microscopic fog from the plates.

(4) Background eradication in nuclear emulsion by the accelerated fading of the latent images.

(1) *Elimination of the development defects introduced by the use of Amidol developers* :

Decrease in the development temperature gives rise to development defects, which are evidenced by a total or partial destruction of the developed image and also a coloration, extending to a considerable depth in the emulsion.

By a series of trials carried out by the author, it was found necessary to increase the ratio sodium sulphite/Amidol for the development temperatures from 18°C to 24°C.

It is an accepted fact that a decrease in the development temperature minimizes distortion in nuclear emulsions. Monothermal development suggested by Yagoda (1955) and Marguin (1957) in which temperature variations are replaced by P. H. variations, was avoided because of its complexity of operation. To avoid distortion, the development was, therefore, carried out at lower than usual temperatures which in turn gave rise to coloration defect, besides an irregularity in development.

To obviate this difficulty, four types of developers as indicated below were tried by the author.

TABLE I

Developer compositions : (in gms/lit. of the solution)

gms/lit. potassium bromide (KBr)	1.2	1.2	1.2	1.2
gms/lit. sodium sulphite (Na_2SO_3)	11	10	17	20
gms/lit. Amidol	4.0	2.8	2.8	2.8
gms/lit. boric acid (H_3BO_3)	30	20	20	20
Ratio of sodium sulphite : Amidol	2.75	3.57	6.07	7.14

By a comparative study of these formulae it was found that to develop reliably at low temperatures, it was necessary to increase the ratio sodium sulphite/Amidol to minimize the coloration. In certain cases of low sodium sulphite concentration, it was observed that the emulsion showed zones of good development, whereas the rest of the plate became useless. Further, in such cases the images were found to show a blue or red transparency. The development was carried out at temperatures higher and lower than 18°C and the coloration was found to be more predominant at lower temperatures, as well as to be a function of development time.

Results, almost resembling those indicated above, were reported by Birge (1954) and others. In some cases the images were completely destroyed,

but one thing is clear that this image destruction has nothing to do with the corrosion which sets in during prolonged fixation, and is more or less a surface phenomenon. In the present case, the disappearance of the image begins deep down in the emulsion.

These phenomena as already reported by James and Vonselow (1953) are due to the displacement of the adsorption equilibria of organic compounds and sodium sulphite, on silver halide grains. The following equation as suggested by Chateau (1956) can explain the phenomenon

$$2Ag\ NaSO_3 \rightarrow 2Ag + Na_2S_2O_6$$

$$AgS_2O_3^- + Ag^+ + H_2O \rightarrow Ag_2S + H_2SO_4$$

All the three sets of plates of 100, 200 and 400 micron thickness were tried, and it was found that all the three sets showed the coloration defect; but the thicker ones are less coloured than their thinner counterparts.

In conclusion, it was found necessary to choose a sodium sulphite to Amidol ratio which is 75% greater than the ratio adopted in Brussels or Chicago developers. Further, the following processing conditions used by the author were found to give best results :

TABLE II

Emulsion thickness (microns)	Water pre-soak		Developer pre-soak		Development time	Stop bath 1% CH_3COOH	
	Temp. T°C	Time Hrs.	Temp. T°C	Time Hrs.		Temp. T°C	Time Hrs.
100	4	0.4	4	0.4	Variable	4°C	0.4
200	4	1.0	4	1.0	Variable	4°C	1.0
400	4	2.6	4	2.6	Variable	4°C	2.6

(Microns)	Fixation (40% hypo)		Washing		Glycerinization One percent	
	Temp. T°C	Time Hrs.	Temp. T°C	Time Hrs.	Temp. T°C	Time Hrs.
100	18	3.5	8	4.0	6	0.5
200	18	8.0	8	10.0	6	1.0
400	18	24.0	8	28.0	6	2.0

Fog was removed by rubbing immediately after the stop bath treatment.

2) *Dependence of track and grain structures on the hot stage temperature, hot stage time and sodium sulphite concentration* :

In this connection various parameters, viz., diameter of the track grains, volumetric grain density, total gap length, mean gap length and blob density

were studied by the author, in the case of Ilford C_2 200 micron plates, some work has already been reported by Fatzer (1959) in this connection. Sodium sulphite/Amidol ratios and temperatures different from those used by Fatzer have been used. Results, although identical with those reported by Fatzer, differ in the peak values of the curves as reported :

a) Diameter of the track grains :

Mean grain diameters were plotted against the development time, for two temperatures and two sodium sulphite concentrations. The results obtained are plotted in Figs. 1 and 2.

Mean diameter of the track grains was found to increase rapidly with the time, in the region of under-development. It, however, attained a constancy in value for longer development periods (Figs. 1 and 2).

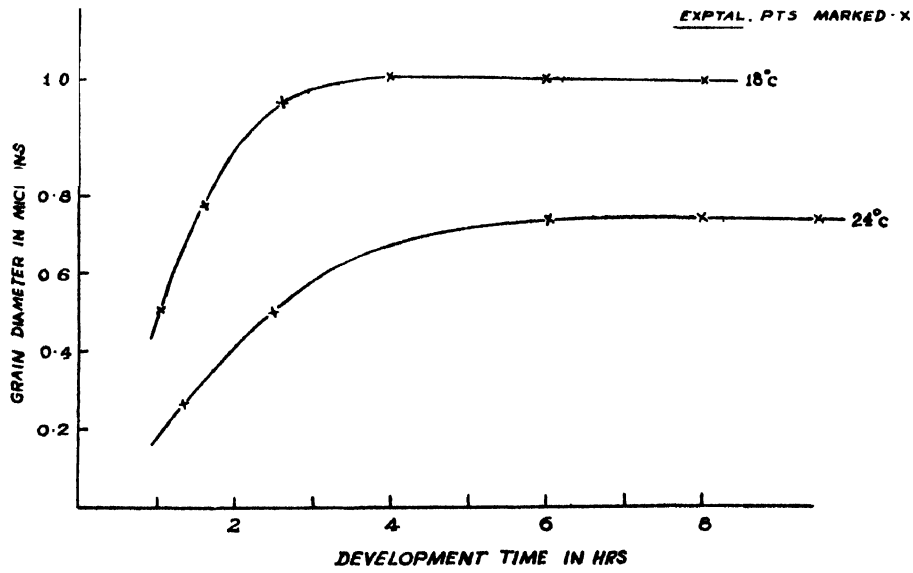

Fig. 1. Grain diameters plotted against development time for two different temperatures.

$(\phi)\frac{\text{Sod. sulphite}}{\text{Amidol}}$ 10

Further, it was obsered that an increase in the sodium sulphite concentration and development temperatures causes a decrease in the grain diameters.

b) Volumetric grain density :

Volumetric grain density was found to increase with an increase in the hot stage time and the sodium sulphite concentration. The observations are plotted in Fig. 3.

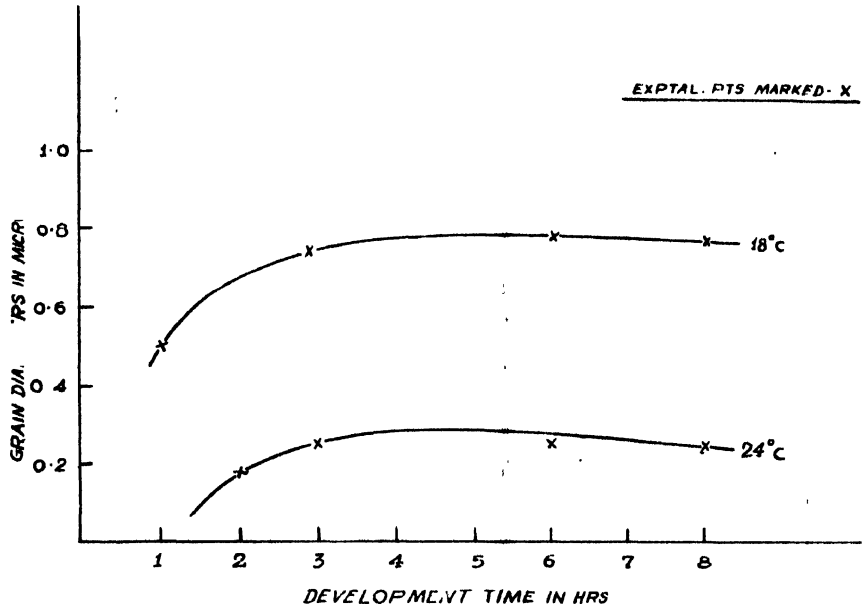

Fig. 2. Grain diameters plotted against the development time at two different temperatures.

$(\phi)\frac{\text{Sod. sulphite}}{\text{Amidol}} = 15$

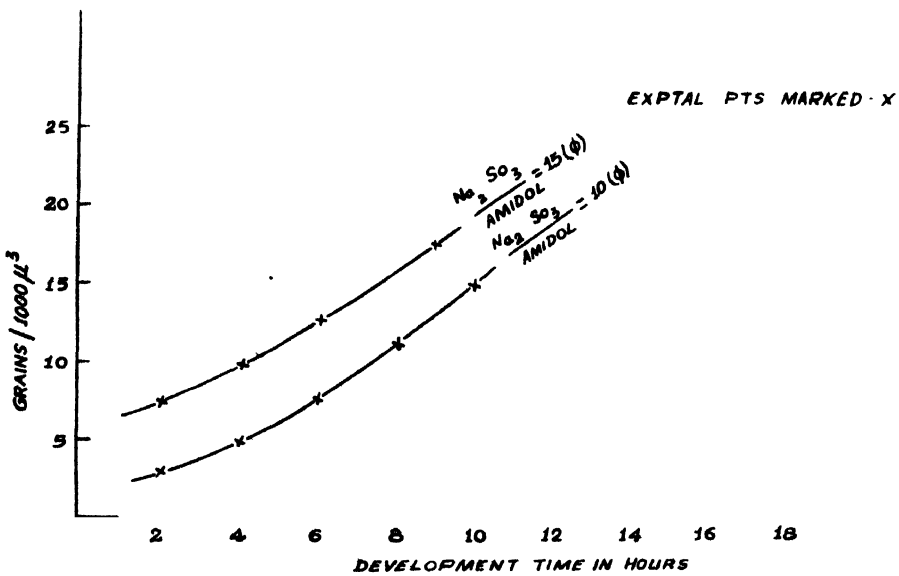

Fig. 3. Volumetric grain density plotted against hot stage time at two Na_2SO_3 concentrations.

1) *Total gap length* :

In the region of under-development total gap length was found to decrease with an increase in the development time. At longer development periods, the total gap length becomes constant. At normal developments, however, the total gap length decreases with increasing temperature and is independent of sodium sulphite concentration. (Fig. 4).

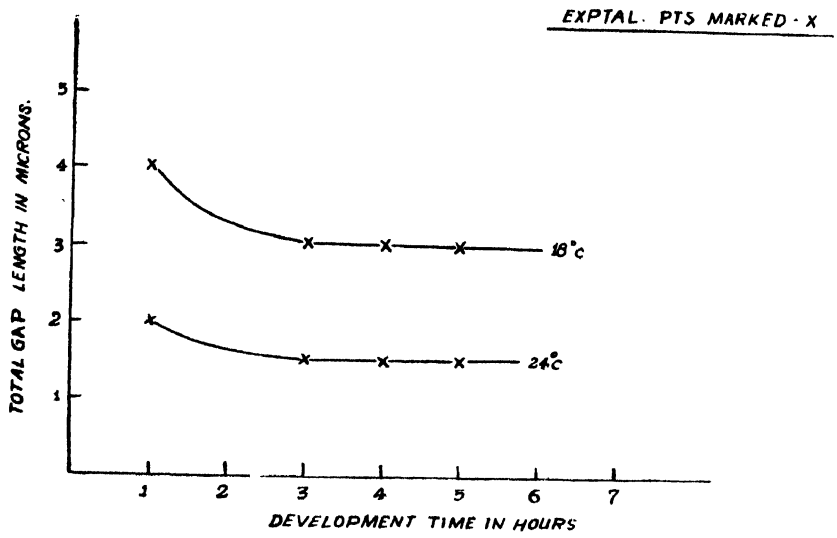

Fig. 4. Total gap length plotted against development time in hours.

2) *Mean gap length* :

The following points were observed in connection with the mean gap length, which was calculated by dividing the total gap length by the number of blobs.

1) M.G.L. is independent of the duration of hot stage time, excepting in the case of under-development.

2) M.G.L. decrease with an increase in temperature.

3) It does not vary with a change in sodium sulphite concentration.

5) *Variation of blob density.*

Blob density was calculated by dividing the number of blobs by the total length of the tracks. It was observed that the blob density increases in the region of under-development. Rise in temperature was found to cause a considerable rise in the blob density and after a certain value of development time, the blob density attained constancy in value. These observations are incorporated in Fig. 6.

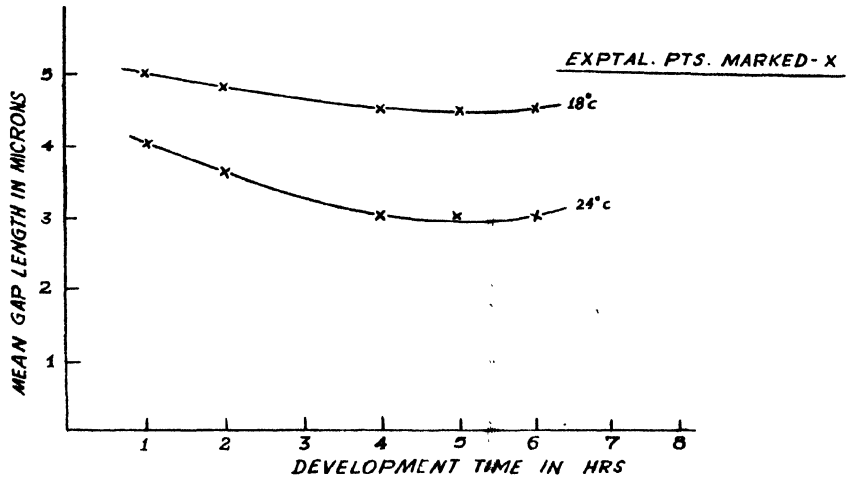

Fig. 5. Mean gap length plotted against the development time at two different temperatures.

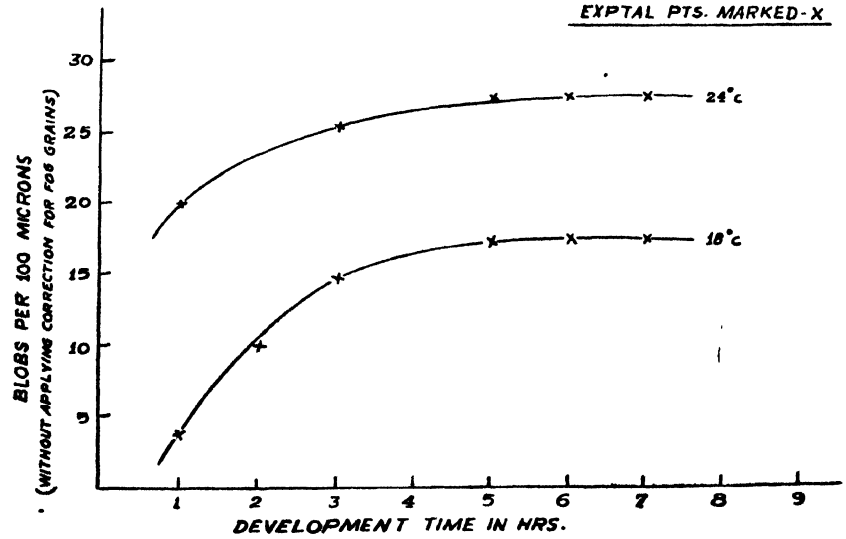

Fig. 6. Blob density plotted against development time in hrs.

(3) *Removal of microscopic fog from the plates* :

Background of developable but unexposed grains of silver bromide, manifesting itself in the form of microscopic fog, is a great handicap in the emulsion work. The idea underlying removal of fog is to develop an unexposed plate to reduce all the developable silver bromide grains, to free silver, which is then removed by reduction. Photographic reducers consist of oxidising agents which

oxidise free silver to a suitable salt, which in turn causes a reduction in the opacity of the image.

Well known formulae using potassium permanganate and potassium bichromate were used in this laboratory. Potassium permanganate formula was found to give better results, and also it was observed that potassium bichromate formula does not cause complete reduction and the grains thus reduced partially are again developable.

Out of the formulae suggested by various workers, the following formula modified by the author was found to give best results, without affecting silver bromide in the emulsion.

Sol. 1—Potassium permanganate— 3 gms water to make
1000 cc
Sol. 2—Sulphuric acid— 8 cc water to make
1000 cc.

Procedure :

The plate was first developed in the Eastman developer *D*–11 for a time greater than the usual, so as to be sure about the development of undesirable grains. The plate was then washed and immersed for about 40 minutes in a freshly prepared mixture of equal volumes of solutions 1 and 2. This procedure was tried for plates of various thicknesses. Following timings were found necessary:

Plate thickness (microns)	100	200	400
Time in minutes	30	40	55

Thus longer time was needed for the clearance of thick plates, as compared to their thinner counterparts. In order to remove the brown stain produced by potastium. permanganate, the plate was again washed and placed for 15 minutes in a 10% solution of sodium bisulphite. This procedure was found best for the removal of the background fog.

4) *Background eradication in nuclear emulsion*

The background is partly due to the accumulated latent images of α tracks and stars produced by traces of Ra and Th, normally present as impurities in the emulsion and partly due to the cosmic ray background.

It was found that storage for several hours in humid atmosphere goes a long way in accelerating the fading of latent images due to the background. The emulsions thus treated were then dessicated for 1 to 2 hrs. over calcium chloride.

Further, the effect of both the temperature and humidity on fading was investigated so as to find out their effect on the eradication of the background tracks.

Effect of temperature on fading :

The variation of the fading rate with the storage temperature may be determined from a consideration of the effect of temperature on the velocity of a gas solid chemical reaction, such as is assumed to occur during fading between certain constituents of the atmosphere and the silver development centres of the exposed emulsion. This gives a form of the Arrhenius relation with $-dN/dt$, the rate of disappearance of the development centres, as the equation

$$\frac{dN}{dt} = Ce^{K/T}$$

where C and K are constants and T the absolute storage temperature. The equation shows that the fading produced under otherwise fixed conditions will be an exponential function of the reciprocal of the absolute temperature, a result confirmed by Farragi (1949). This shows the effect of storage temperature on the background eradication and it is found that the experimental findings are in accord with the theory.

Effect of humidity on fading:

According to Albouy and Farragi (1949) fading is a result of the oxidation of the development specks by the atmospheric oxygen in the presence of water, the reaction as proposed by Albouy and Farragi proceeds as

$$Ag+O+H_2O \rightarrow Ag+2OH^-$$

It is evident that the presence of an excess of OH^- ions i.e. PH value above 7, will act to inhibit the reaction, while the more acid conditions accelerate it. Thus, an increase in the humidity causes a decrease in the fading rate.

The following graph showing the effect of both temperature and humidity on the background eradication is in accord with the theory.

Theoretical investigations due to Beiser (1951) also lead to the following equation for the fading coefficient

$$\frac{D_0 - D}{D_0} = 1 - \text{Exp.}(-Ct)$$

where D_0 = grain density produced upon immediate development

D = grain density after a time t

C = constant depending upon the size of Ag specks

The fading coefficient plotted as a function of t, for various values of C, is in agreement with the experimental curves.

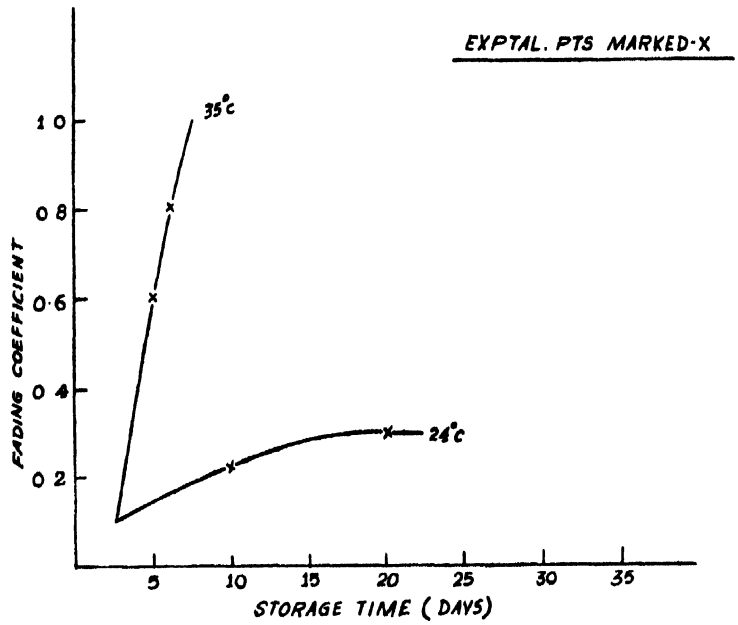

Fig. 7. The variation of the fading coefficient with the time of storage at various temperatures.

ACKNOWLEDGMENTS

The author expresses his deep sense of gratitude to Prof. B. D. Nag Chaudhuri, under whose direction and guidance this work has been carried out. The author is also thankful to Mr. S. K. Mukherjee for providing the laboratory facilities ; and to Messrs. Naresh Sen and Barin Chatterjee for their valuable assistance.

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